U. S. Army

Chemical Research and Development Laboratories

Technical Report

CRDLR 3085

Free-Fall Breakup of Bulk Liquids

by
James D. Wilcox
Joseph V. Pistritto
Alan B. Palmer

September 1961

ARMY CHEMICAL CENTER, MD.

September 1961 CRDLR 3085

FREE-FALL BREAKUP OF BULK LIQUIDS

by

James D. Wilcox
Joseph V. Pistritto
Alan B. Palmer

Physicochemical Research Division

Recommending Approval:

Carl M Herget

CARL M. HERGET, Ph.D.
Director of Research

Approved:

S. D. SILVER
Scientific Director

U. S. ARMY
Chemical Corps Research and Development Command
CHEMICAL RESEARCH AND DEVELOPMENT LABORATORIES
Army Chemical Center, Maryland

FOREWORD

This work was conducted under Task 4C08-03-016-10, Agent Physicochemical Research (U), Subtask 2, Aerosol Studies (U). The work was started in September 1957 and completed in February 1959. The data are recorded in notebook 4878.

Notice

Disposition

When this document has served its purpose, DESTROY it. DO NOT return the document to U. S. Army Chemical Research and Development Laboratories.

DIGEST

In this study the mechanism and fundamental characteristics of the breakup of large masses of liquid under free-fall conditions were investigated in relation to the high-altitude release of liquids from a container for the purpose of forming aerosols.

This report describes the bulk-liquid breakup of 12.5-ml to 2.5-l volumes of water and 25-ml volumes of dibutyl phthalate and dibutyl phthalate solutions containing 0.5% and 2% polyvinyl acetate, a high-molecular-weight polymer. The liquid was released uniformly from a height of approximately 7 m to 8 m, and its breakup under free fall was recorded by high-speed photographic techniques.

The following conclusions were reached:

1. The predominant mode of breakup of a bulk-liquid mass subjected to free-fall conditions occurs by the bursting-bag or canopy-formation process, and

2. Certain physical factors in the liquid mass, such as surface tension, size, initial shape, velocity at the time of release, and concentration of high-molecular-weight polymer, have a modifying effect on its breakup.

CONTENTS

FREE-FALL BREAKUP OF BULK LIQUIDS

I. INTRODUCTION.

The mechanism and fundamental characteristics of the breakup of large masses of bulk liquid are being studied in relation to high-altitude release of liquids from a container. The container might be a bomb, projectile, or tank released from an aircraft, guided missile, balloon, flying platform, etc. This report covers the bulk-liquid breakup of water and other liquids under free-fall conditions.

Extensive literature surveys by Armor Research Foundation[1, 2] have revealed no previous experimental or theoretical studies of the disintegration of bulk liquids under free fall; therefore, a fundamental research study of this problem was undertaken. It was proposed that this study consist of bulk-liquid breakup under free-fall conditions and at velocities of up to 500 ft/sec. The breakup of a liquid mass is governed by either its terminal velocity or its velocity at the time of release, whichever is greater. This velocity factor dominates all other factors involved in the breakup of a liquid mass.

Liquid breakup is caused by instability. All Newtonian liquids greater than a certain critical size are unstable during free fall. Some important factors known to affect drop breakup or stability are:

1. Drag coefficient
2. Surface tension
3. Acceleration
4. Liquid volume
5. Liquid velocity
6. Liquid density
7. Liquid viscosity
8. Air density
9. Air viscosity

Factors affecting the stability and breakup of liquid drops are also given in a theoretical study of the breakup of liquid masses by the canopy-formation method.[3] This theoretical study was undertaken as a part of this mass-breakup study.

The end results of the physical processes with which this study is concerned are the breakup elements and the breakup products. Their physical characteristics, e.g., mass, geometrical shape and size, and velocity of fall, are of interest, not only on their own account, but because they indicate the nature and intensity of the processes involved in the breakup.

For example, a study of the breakup of a mass of liquid during its fall towards the ground may throw some light on how the liquid separates into the different stages of the breakup process; i.e., how the drops distribute themselves, how they coalesce, what factors influence the breakup, and what the ultimate particle-size distribution will be. Ideally, a detailed theory of liquid breakup should account for the final particle-size distribution and its variation in space and time, and predict the intensity with which the particles fall. Some knowledge of the terminal velocities of the breakup elements as a function of their mass and shape is highly desirable.

Although there is little direct observational evidence, there is good reason to believe that the size distribution of a mass of liquid must change continually in its fall toward the ground. After maximum breakup, effects are still produced by the coalescence between drops, the separating effects of wind shear and gravitational settling, and the evaporation of the drops. There are no data available on how concentration and size distribution vary in time and space.

Bulk-liquid breakup occurs in two stages: first, the breakup of the liquid into drops; and second, the fall of the drops. The breakup of the liquid into drops is covered experimentally in this study.

II. HISTORY.

A. Prior Velocity Studies.

The fall of drops has been of interest to investigators for many years. In 1904, Lenard[4] first measured the falling velocity of waterdrops of raindrop size. In 1909, Schmidt[5] measured the falling velocity of raindrops of 0.4 mm to 3.5 mm. More recently, in 1941, Laws[6] and, in 1949, Gunn and Kinzer[7] investigated a range of diameters from 8μ to 5.8 mm to obtain their fall velocities. They showed that the fall velocities given by Lenard and Schmidt were about 15% low.

Laws measured the velocities of 1-mm- to 6-mm-diameter waterdrops in still air from heights of 0.5 m to 20 m. Drops of up to 6 mm in diameter reached 95% of their terminal velocities after falling less than 8 m, and for all practical purposes could be said to reach their full terminal velocities after falling 20 m. Laws found that 6-mm raindrops had slightly lower terminal-velocity values than those determined experimentally for 6-mm drops in still air after 20 m of fall. Laws attributed these lower terminal-velocity values for raindrops to small-scale turbulent motions in the atmosphere. It has been assumed that, since drops of a diameter greater than 6 mm are rarely observed in rain, drops larger than this usually break up during their fall to the ground.

It would be desirable to have a formula for the terminal velocity of a drop of any size falling through air of any density. There is little hope, however, of deriving a reasonably simple expression to account for all of the factors involved; e.g., shape, circulation effects, etc. In 1942, Davies[8] prepared an equation that is reported to predict within 3% the terminal velocities of drops of any liquid in air at pressures down to 0.5 atm. In 1950, Best,[9] using work of Davies, also prepared equations to predict the terminal velocities of liquid drops of various sizes. In 1958, Repp and Magram[10] prepared an excellent paper in which they gave empirical equations for terminal settling velocities. The terminal velocities of waterdrops as found by several investigators are given in table 1.[6,7,11,12]

B. Prior Drop-Shape Studies.

Drops exist as mechanically stable systems because the surface forces at the liquid-air interface continually try to minimize the interfacial area. When the drops are small enough, as in the case of cloud droplets, drizzle, and small raindrops, the surface-tension forces are predominant and the shape is one of minimum surface-to-volume ratio; i.e., a sphere. When factors other than surface energy contribute significantly to the total energy of a drop, however, minimum total energy may become inconsistent with a spherical shape.

In 1954, Magono[13] showed that large waterdrops falling at terminal velocity do not possess the streamlined or teardrop shape but exhibit a marked flattening on their lower surfaces and a smoothly rounded curvature on their upper surfaces.

TABLE 1

TERMINAL VELOCITIES OF WATERDROPS IN ATMOSPHERE

Diameter	Velocity			
	Mason[11]	Gunn and Kinzer[7]	Laws[6]	Drinker and Hatch[12]
μ	cm/sec			
20	1.3	—	—	—
30	3.0	—	—	—
40	5.4	—	—	—
50	—	—	—	7.5
65	11.3	—	—	—
80	18.3	—	—	—
100	26.5	27	—	30
200	76	72	—	—
300	125	117	—	—
400	172	162	—	—
500	217	206	—	282
600	260	247	—	—
700	302	287	—	—
800	344	327	—	—
900	386	367	—	—
1,000	426	403	—	401
1,100	462	—	—	—
1,200	493	464	—	—
1,250	—	—	485	—
1,400	551	517	—	—
1,500	—	—	551	—
1,600	601	565	—	—
1,750	—	—	608	—
1,800	645	609	—	—
2,000	690	649	658	—
2,250	—	—	702	—
2,400	769	727	—	—
2,500	—	—	741	—
2,750	—	—	776	—
2,800	825	782	—	—
3,000	—	806	806	—
3,200	870	826	—	—
3,250	—	—	831	—
3,500	—	—	852	—
3,750	—	—	871	—
4,000	925	883	886	—
4,500	—	—	910	—
4,800	955	907	—	—
5,000	—	909	925	889
5,500	—	—	930	—
5,600	970	916	—	—
6,000	976	—	930	—

In 1954, McDonald[14] showed that the effects of electrostatic charges and internal circulations within a drop are of negligible importance in controlling its shape. He concluded that the aerodynamic pressures and the surface-tension-pressure increments combine to produce an internal-pressure pattern that satisfies the hydrostatic equation. If the surface geometry of a drop is known, the surface-tension- and hydrostatic-pressure differences at any point can be calculated. After these pressure differences are known, it is possible to compute the aerodynamic-pressure profile along a meridian if the fall velocity of the drop is known. McDonald's hypothesis of the balance of surface-tension-, hydrostatic-, and aerodynamic-pressure forces agrees with experimental evidence.

In 1949, Gunn[15] interpreted his findings regarding the tendency of waterdrops 1 mm in diameter to sideslip as they fall as the resonance between the natural frequency of mechanical oscillation of the drops and the frequency with which eddies are shed from the upper surface of the drops.

In 1950, Blanchard[16] also observed the periodic oscillation of waterdrops moving in a vertical airstream. He was able to suspend successfully 9-mm drops of water in a nonturbulent airstream.

In 1951, Lane[17] allowed drops to fall in an airstream and photographed their breakup. The drops first flattened and formed a circular ring with a thin membrane in the center. This membrane then blew out into a hollow bag that burst, producing a shower of fine droplets.

In 1956, Magarvey and Taylor[18] in their experiments with drops 10 mm to 20 mm in diameter, showed that the instability and breakup of large drops were the same as those shown by Lane with smaller drops.

In 1952, Hughes and Gilliland[19] made an excellent mathematical review and analysis of the mechanics of drops.

The rigorous theoretical analysis by Hinze[20] on the forced deformation of liquid globules indicates that there is a certain critical Weber number to be used in predicting the breakup of a liquid globule. The Weber number is a function of surface tension.

In 1947, Merrington and Richardson[21] experimentally determined that the critical Weber number of a splitting liquid drop was about 10.

These drop-breakup studies by previous investigators are quite comprehensive, and the results of these investigations will be used in this study. As stated previously, therefore, this experimental study covers the breakup of large masses of liquids into drops.

III. EXPERIMENTATION.

In this study of the free-fall breakup of bulk liquids, it was decided that quantities of liquid varying from 25 ml to 2.5 ℓ could be conveniently handled in the laboratory.

A. Still-Photography Method.

Experimentally, it was desirable to know the velocity of the falling liquid or its breakup products at any and all distances. Provisions were made, therefore, so that the time after release of the liquid and its distance of fall could be measured. High-speed photographs of the fall of the liquid should give information on the process of the liquid breakup.

1. Liquid-Release Mechanism.

The liquid-release system had to be able to release the liquid suddenly from its container and subject it to free fall. The liquid-release process had to be reproducible and cause little or no disturbance to the liquid itself. The release mechanism consisted of a cylindrical glass tube with a 1-inch inside diameter, over which a thin rubber diaphragm was stretched. The cylindrical glass tube with the diaphragm in place on the bottom end acted as a container for the liquid. A strand of no. 30 copper wire was placed externally across the diaphragm. This wire exploded when it was suddenly subjected to a large current flow. The exploding wire caused the sudden rupture of the rubber diaphragm, which in turn released the liquid from the cylinder. This method of liquid release was very reliable for 12.5-ml and 25-ml volumes.

2. Time-of-Fall and Distance-of-Fall Measurement Methods.

Figure 1 is a block diagram for the time-delay and release-mechanism systems. In essence, a switch was closed, causing the breaking of the diaphragm and the release of the liquid. The closing of this switch also sent a simultaneous pulse to an oscilloscope previously set for a predetermined time delay. After this delay, a pulse was given off by the oscilloscope, firing the camera shutter, which in turn was synchronized and fired the microflash unit. The delay set on the oscilloscope represented the time of fall of the liquid

or, more accurately, the time from the bursting of the diaphragm until the flash photograph was taken. There was an inherent delay in the relay between the event switch and the exploding wire. There was also another inherent delay in a relay, the camera solenoid, and the camera shutter. These delays were relatively constant and were taken into account in recording the time of fall.

The camera and flash unit remained in a fixed position. The liquid in the release setup was raised to various measured heights above the camera. This technique made it necessary to release a new liquid mass and photograph it for each height selected. It was, of course, necessary to increase the delay time as the liquid was raised to greater heights above the camera. A meter stick was placed in the photographic field to make reasonably accurate measurements of the distances of fall. A small quantity of potassium permanganate was dissolved in the water to aid in photography. The surface tension of the water was not affected by the addition of the potassium permanganate.

Since the liquid was released from a tube, the bottom edge of the liquid, having no cylinder walls to cause a drag on it, fell directly into the open sooner than the upper portion of the liquid. For this reason, the distances involved were measured by referring to the bottom edge of the liquid, rather than to any other portion of the liquid mass. The lower portion of the liquid started falling as soon as the diaphragm was broken; thus, the times were measured from the instant the wire exploded.

3. Method for Calculating Liquid-Fall Velocity.

The distances and times of free fall of the liquid were recorded, and a graphical representation of these data was made in the form of a distance-time plot (figure 2). The fall of the main liquid mass and the fall of the main droplet mass are represented by solid and broken lines, respectively. The velocities of both the main liquid mass and the droplet mass were plotted against distance of fall (figure 3). The data for this velocity plot were obtained by graphically differentiating the distance-time curve.

B. Motion-Picture-Photography Method.

The techniques used in the high-speed still-photography method gave excellent photographic results; however, considerable labor was required to get reliable data. High-speed motion-picture photography would rapidly give information on the time and distance of fall of the liquid as well as photographic information on the breakup of the liquid.

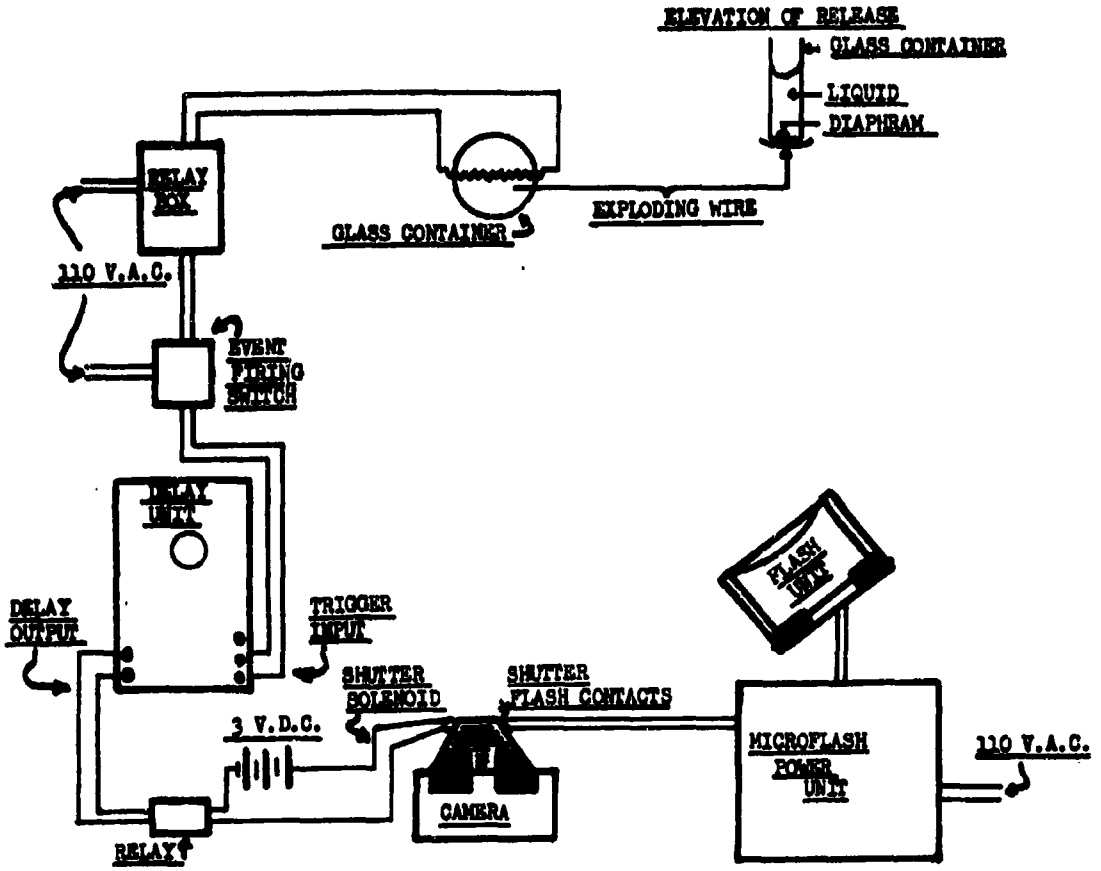


FIGURE 1

BLOCK DIAGRAM FOR TIME DELAY AND RELEASE MECHANISM FOR MASS-LIQUID-BREAKUP STUDY

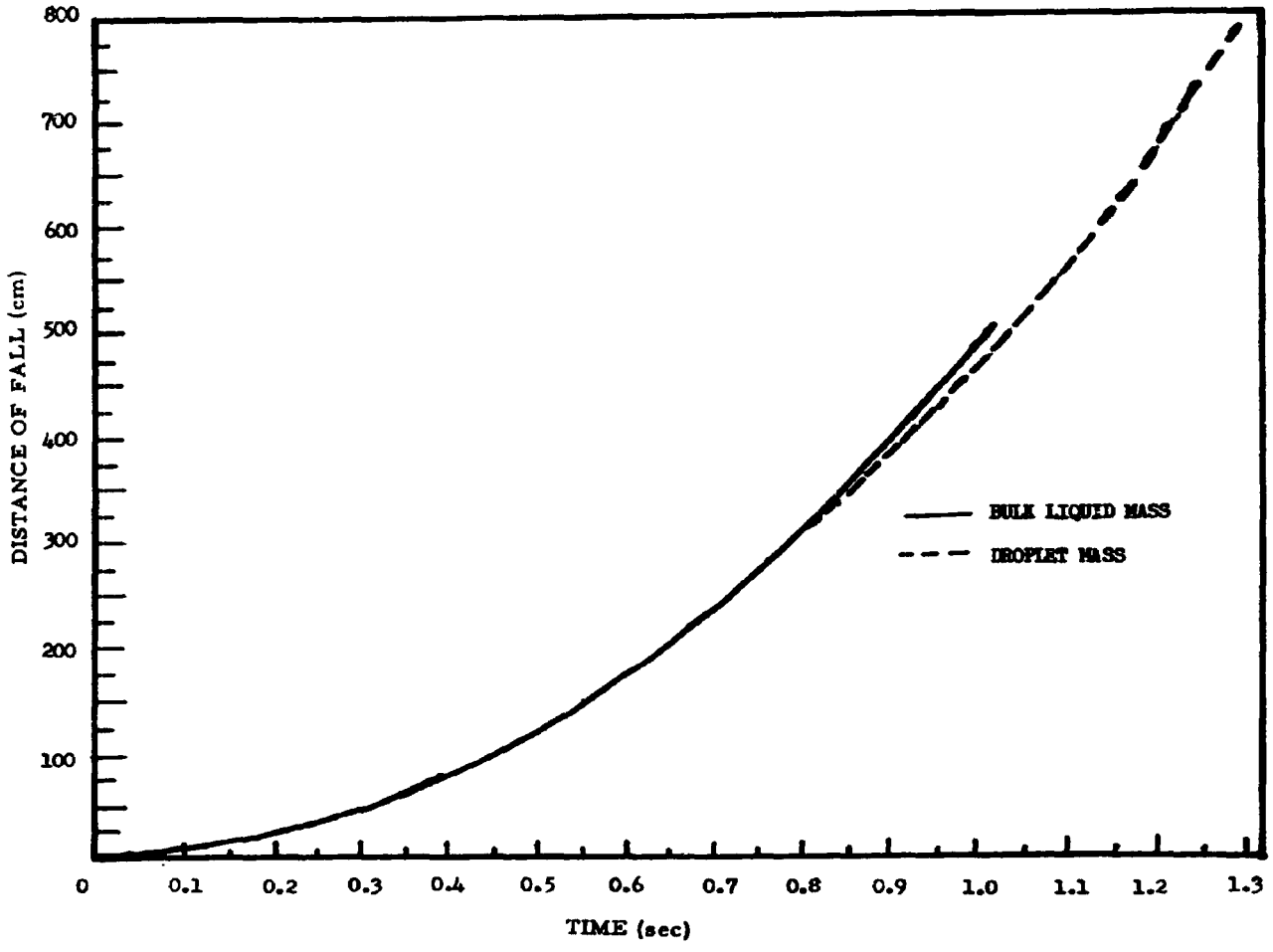


FIGURE 2

DISTANCE-TIME RELATIONSHIP OF RELEASE AND FREE FALL OF 25 ML OF WATER DETERMINED BY HIGH-SPEED STILL-PHOTOGRAPHY METHOD

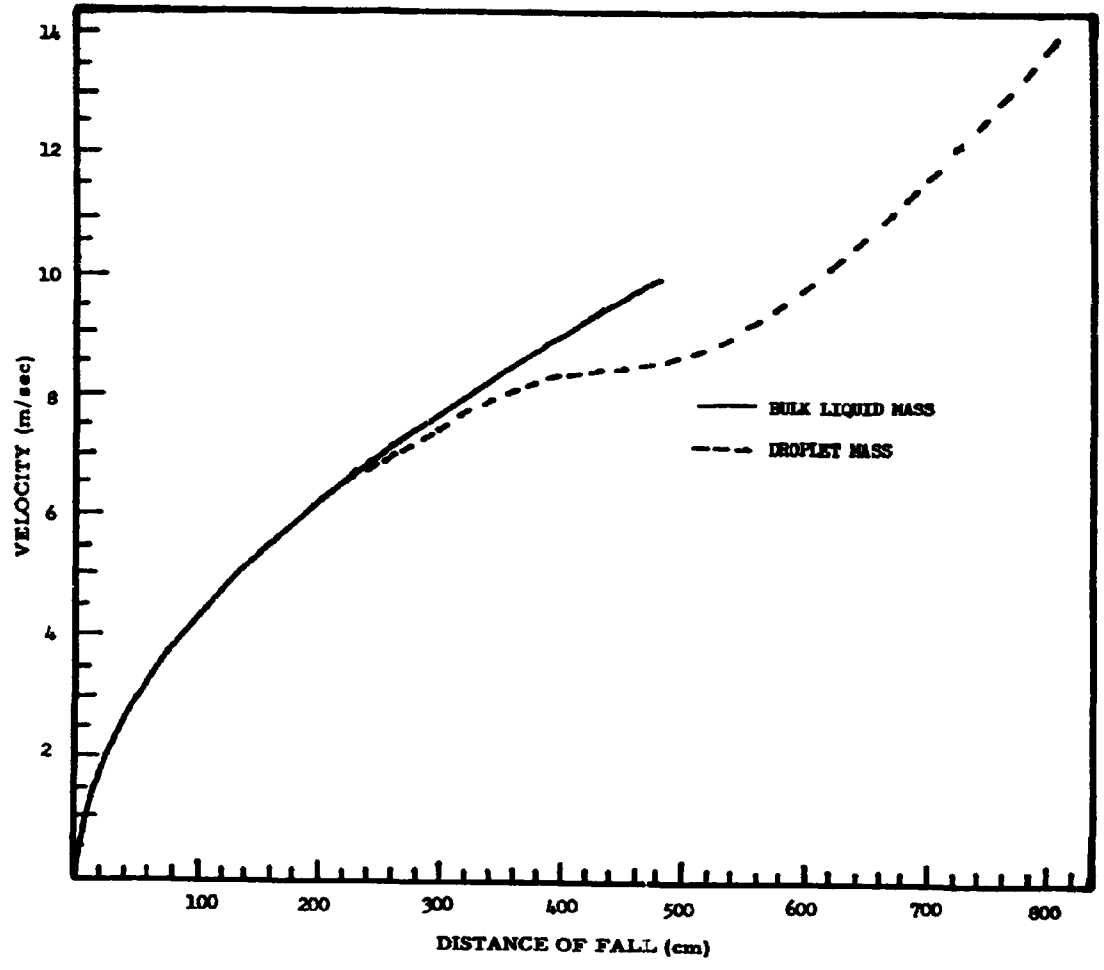


FIGURE 3

VELOCITY-DISTANCE RELATIONSHIP OF RELEASE AND FREE FALL OF 25 ML OF WATER DETERMINED BY HIGH-SPEED STILL-PHOTOGRAPHY METHOD

1. Description of High-Speed Motion-Picture System.

Figure 4 is a block diagram showing the relationship among the different components used in making high-speed motion-picture recordings of the release of a large liquid mass. The camera was a 16-mm Fastax high-speed movie camera, operated at a film speed of approximately 2,800 frames/sec. The sequence followed to cause the fall of the liquid mass and its subsequent recording on film in the test commenced with the starting of the camera to allow the film to reach a desired film speed. After the film had reached the desired velocity, the event was started by a timing mechanism associated with the camera. This time delay was set previously for a specific time delay and completed a battery-operated circuit that closed a relay allowing a 110-v alternating current to pass through a no. 30 copper wire. This current surge caused the wire to explode, initiating the rupture of a thin rubber diaphragm covering the bottom of the cylindrical glass container holding the volume of liquid to be tested. This liquid-release mechansim has been described previously in section III, A, 1.

A slight variation in the liquid-release mechanism was used with the 2.5-ℓ volumes. In the instantaneous release of a large volume of liquid, it was not feasible to discharge or pour the liquid from a bucket or beaker, or to allow the liquid to fall suddenly from an open-end cylinder and still obtain a uniform release pattern or an accurate picture of the sequence of events taking place.

To solve this problem, the investigators filled a thin rubber container with 2.5 ℓ of water, punctured the container with a hypodermic needle, and photographed the breaking of the rubber container and the subsequent release of its contents. An electronic microflash unit having a flash duration of 2 μsec was used to control the exposure time of the photographs in an effort to arrest any action taking place. As a result of the study, these thin rubber containers were used to contain the larger quantities of water used in subsequent tests. In these tests, the rubber container was filled with 2.5 ℓ of water and positioned in place of the cylindrical glass tube used in the 25-ml tests. The stressed-rubber container was ruptured by hot metal particles emanating from an electrical squib fired by a battery. The circuit to the battery and squib was controlled directly by the time-delay unit of the Fastax camera itself.

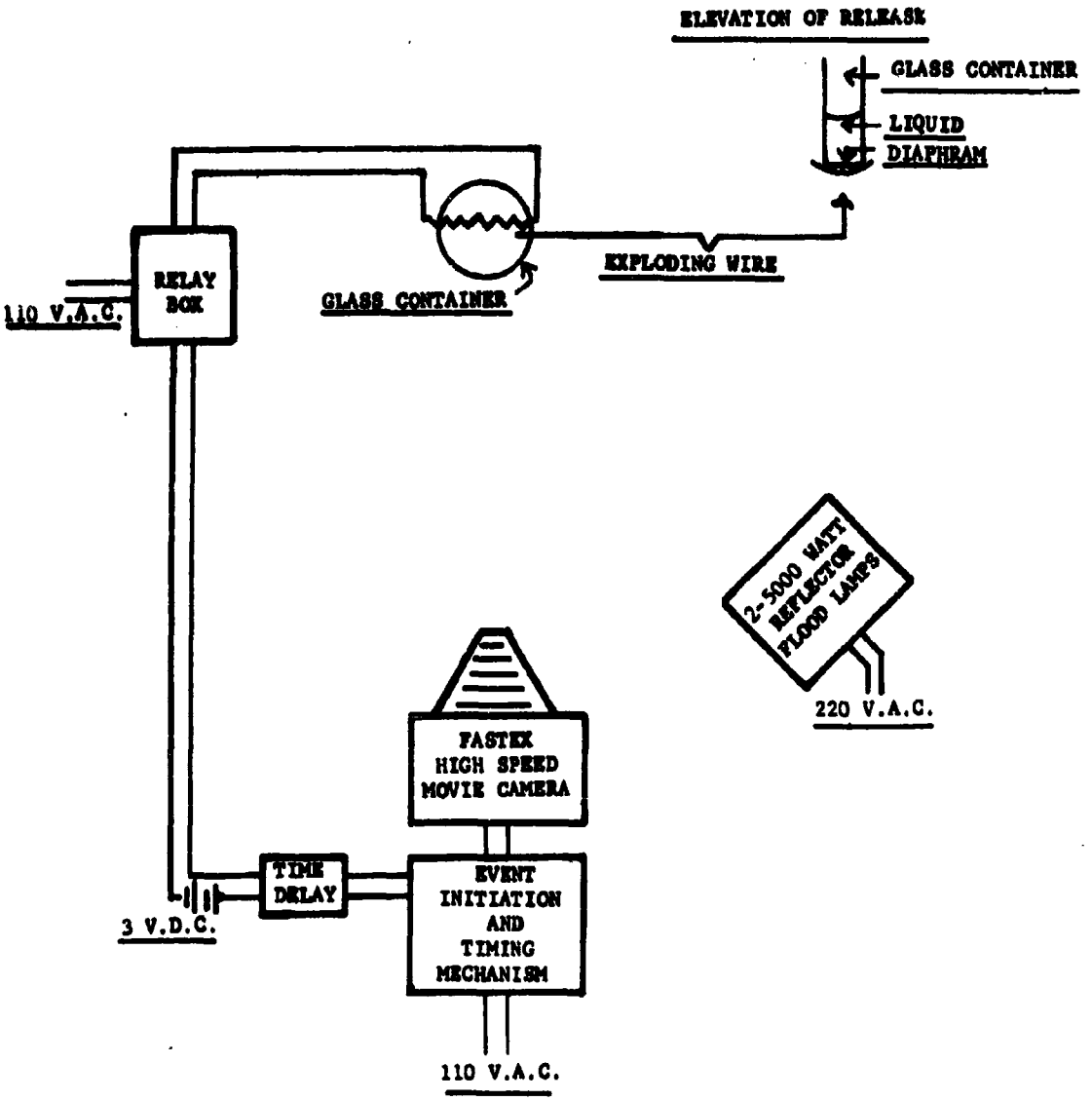


FIGURE 4

BLOCK DIAGRAM OF RELEASE MECHANISM AND HIGH-SPEED-MOVIE RECORDING OF MASS-LIQUID BREAKUP

The light for all of these tests was provided by two, 5,000-w, reflector floodlamps positioned (as shown in figure 4) at a 45° angle with respect to the camera. This angle was determined experimentally and was found to yield the best photographic results.

In view of the restrictions imposed by the optical system of the Fastax camera, it was only possible to record 1 m of fall of the released liquids in any one test. It was therefore necessary to conduct eight separate test trials in increments of 1 m to cover a release distance of 8 m for each liquid tested.

A severe difficulty attributable to the transparent nature of water and dibutyl phthalate, occurred in making these film recordings. It was determined experimentally that the photographic reproduction of the liquid masses could be improved greatly by the addition of extremely small quantities of fluorescent dye materials. In the case of water, a concentration of 0.025 gm of fluorescein sodium per liter of water produced excellent results. In the case of dibutyl phthalate, with and without the viscoelastic additive, a concentration of 0.025 gm/ℓ of Fluorol 7 GA* was sufficient to produce satisfactory results. In the latter case, the quantity of dye added had no measurable effect on the surface tension and viscosity of the dibutyl phthalate. The addition of 0.025 gm of fluorescein sodium to 1 ℓ of water slightly decreased the surface tension of the water. The effect of this decrease in surface tension is discussed later in this report.

2. Free-Fall Breakup Tests.

The liquids used are given below. All liquids contained approximately 0.0025% of fluorescent dye to aid in the photographic recording of the breakup events.

a. Tests Using 25 ml of Liquid.

(1) Distilled water

(2) Dibutyl phthalate

* Fluorol 7 GA is a product of the General Dyestuff Corporation, 435 Hudson Street, New York 14, N. Y.

(3) Dibutyl phthalate containing 0.5% polyvinyl acetate (Lemac 1000*)

(4) Dibutyl phthalate containing 2.0% polyvinyl acetate (Lemac 1000)

b. Test Using 2.5 ℓ of Liquid.

(1) Distilled water

(2) Technical difficulties prevented the testing of additional liquids.

3. Method of Film Analysis.

As stated previously in section III, B, 1, film recordings of the released liquid masses were made in distance-of-fall increments of 1 m. In making these film recordings, a 1-m marker with lines inscribed at intervals of 10 cm was placed in the field of view of the Fastax camera. In this manner, the distance traveled by the liquid mass within this 1-m field of view could be obtained at any specific time by simply observing the position of the leading edge of the liquid with respect to the marker. This distance, plus the distance from the marker to the point at which the liquid was released, would give the total distance traveled by the liquid at any time after release.

The Fastax camera is equipped with a small neon light that flashes at a constant rate of 120 flashes/sec. This light produces a series of dark and light areas along the margin of the entire length of the film as the film travels through the camera. The time interval between the beginning of one dark area and the beginning of the next dark area is 0.00833 sec. The average film speed in this interval, in frames per second, can be obtained by counting the number of individual frames in the interval and dividing by 0.00833 sec. Since these intervals represent a definite time increment, they can be cumulatively summed to obtain the elapsed time at any distance of fall of the released liquid volume. A distance-time plot of these data is shown in figure 5.

* Lemac 1000 is a high-molecular-weight polymer used to impart viscoelastic properties to certain liquids. It is a product of the Borden Co., Chemical Div., 305 Madison Ave., New York 17, N.Y.

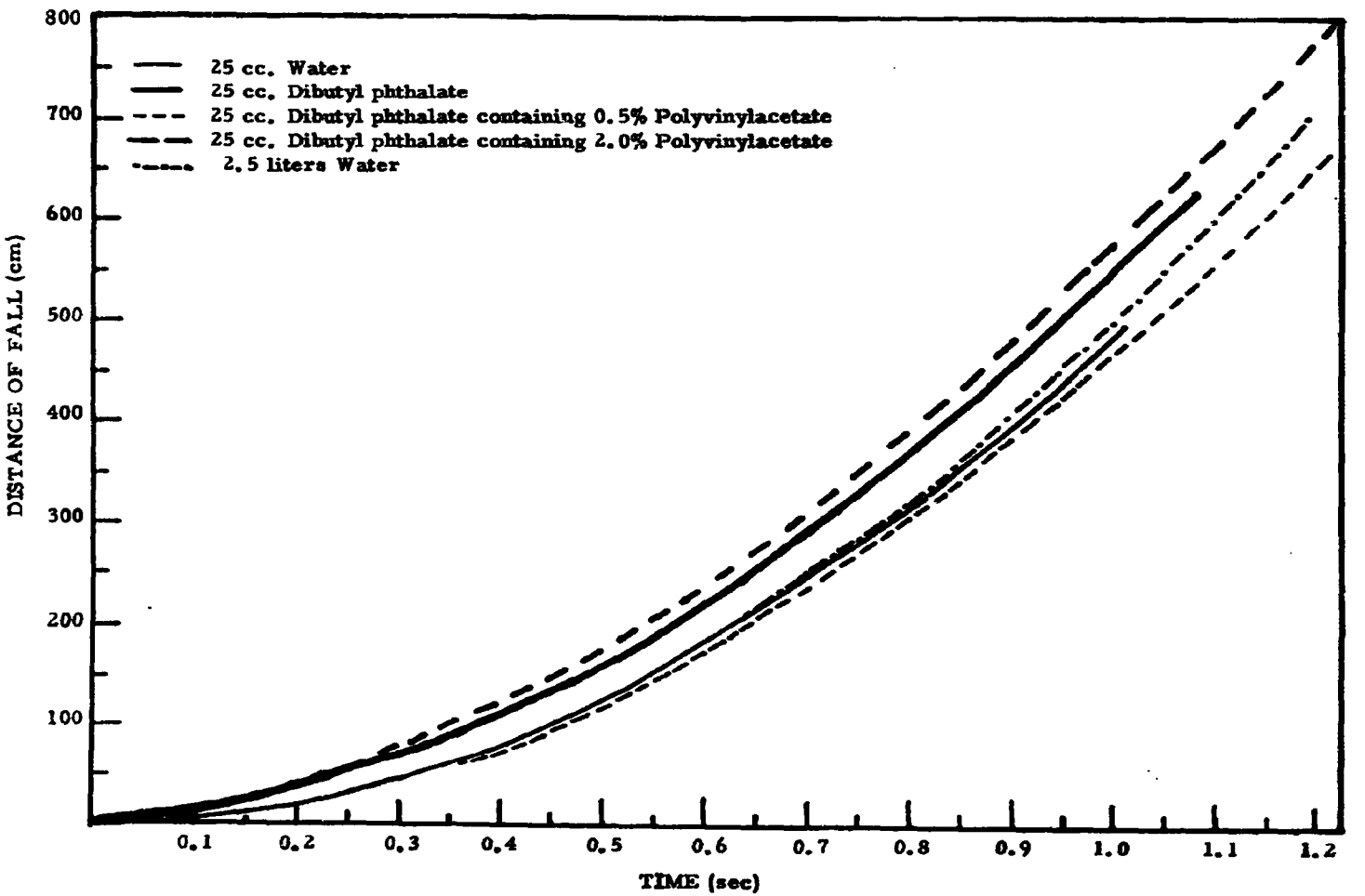


FIGURE 5

DISTANCE-TIME RELATIONSHIP OF FREE FALL OF LIQUID MASSES
DETERMINED BY HIGH-SPEED-MOVIE TECHNIQUE

IV. RESULTS.

A. Still-Photography Method.

Figure 6 is a composite photograph representing the uniform release of liquid from a container after rupture of the rubber diaphragm at the bottom of the glass tube by an exploding wire in three individual tests. In each of the three individual photographs, the liquid has traveled the same distance (2 cm) after an equal time period (0.05 sec). The smoke trails in these photographs were caused by the exploding wires.

Figure 7 is a series of photographs showing the flow of the liquid from the container and the point at which it finally breaks away from the container. The time for complete flow from the container was 0.20 sec. At this time, the liquid had fallen 22 cm and had reached a velocity of approximately 2 m/sec.

In this part of the investigation, both 12.5-ml and 25-ml volumes of water were studied. With the 12.5-ml volumes, the height of the column of liquid was about the same as its width (1 inch); with the 25-ml volumes, its height was approximately twice its width. In the tests with the smaller volumes, only one initial breakup pattern was observed. With the larger volumes, however, the initial breakup pattern repeated itself (figures 8 and 9).

Figure 8 is a series of photographs showing the initial breakup of 25 ml of water in free fall. The liquid flattened out into an inverted saucer-like shape in 0.40 sec. The distance of travel for this flattening to take place was about 75 cm; the liquid had attained a velocity of about 4 m/sec. The liquid then began to develop a baglike appearance as in Lane's[17] drop-breakup investigations. After falling approximately 2.5 m and reaching a velocity of approximately 7 m/sec, 0.70 sec after release, the bag broke, producing a spray of smaller drops.

Figure 9 is a photograph of the subsequent breakup of the 25-ml volume of water. This second bag formed and broke very rapidly at a distance of about 3 m (0.80 sec after release), having reached a velocity of approximately 8 m/sec. The remaining small volume of original water contained in the ring of the second broken bag continued to break up, and no original water volume was observed after approximately 5 m of fall. After 5.5 m of fall, the droplet volume had attained a velocity of about 9.5 m/sec. The maximum distance of fall for this investigation was physically limited to 8 m. The time of fall from release to a fall of 8 m for the liquid was 1.3 sec, and the droplet-mass velocity was approximately 14 m/sec at the 8-m point.

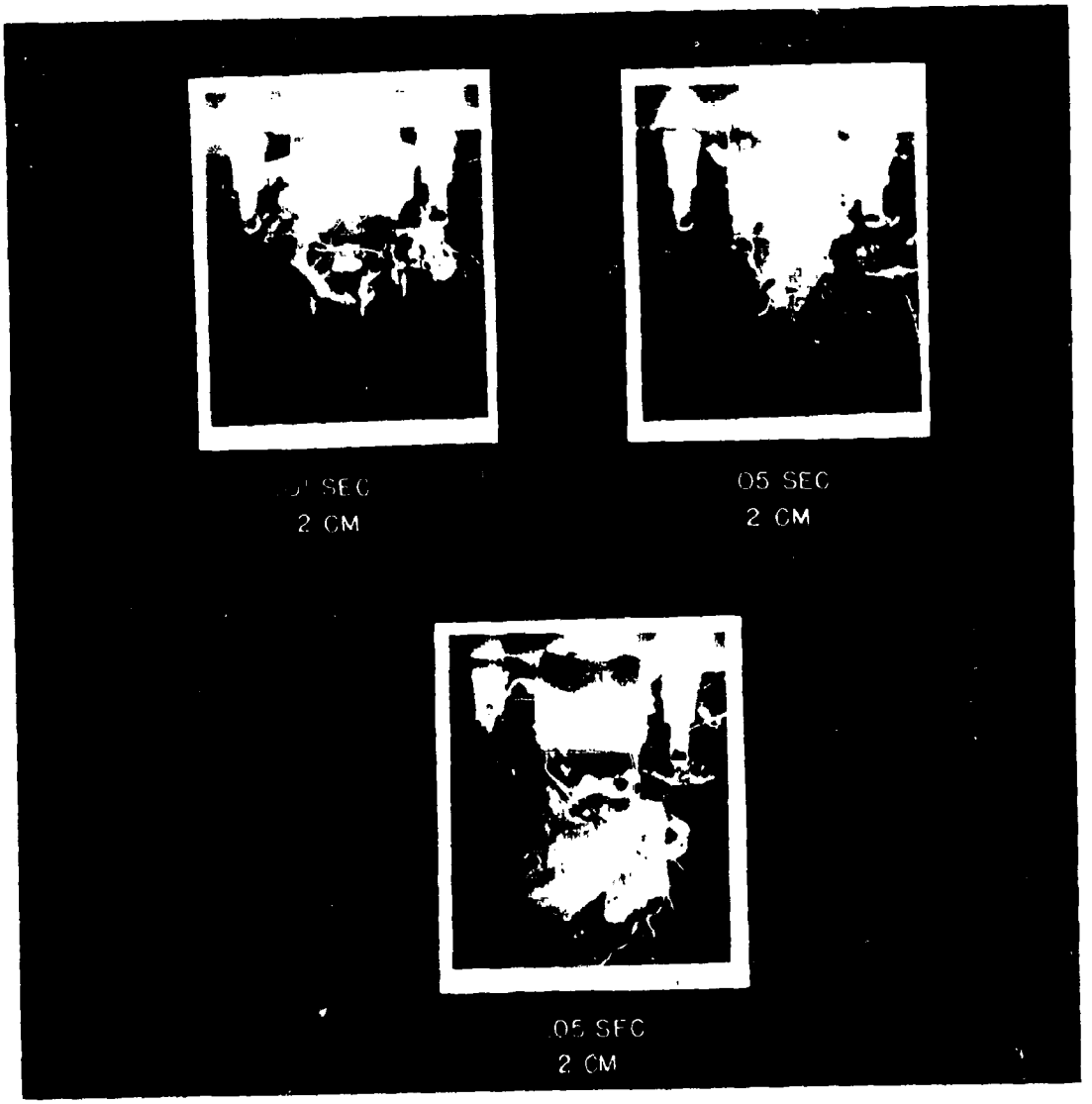


FIGURE 6

UNIFORM RELEASE OF WATER — TIMES AND DISTANCES
OF LIQUID FALL AFTER DIAPHRAGM RUPTURE

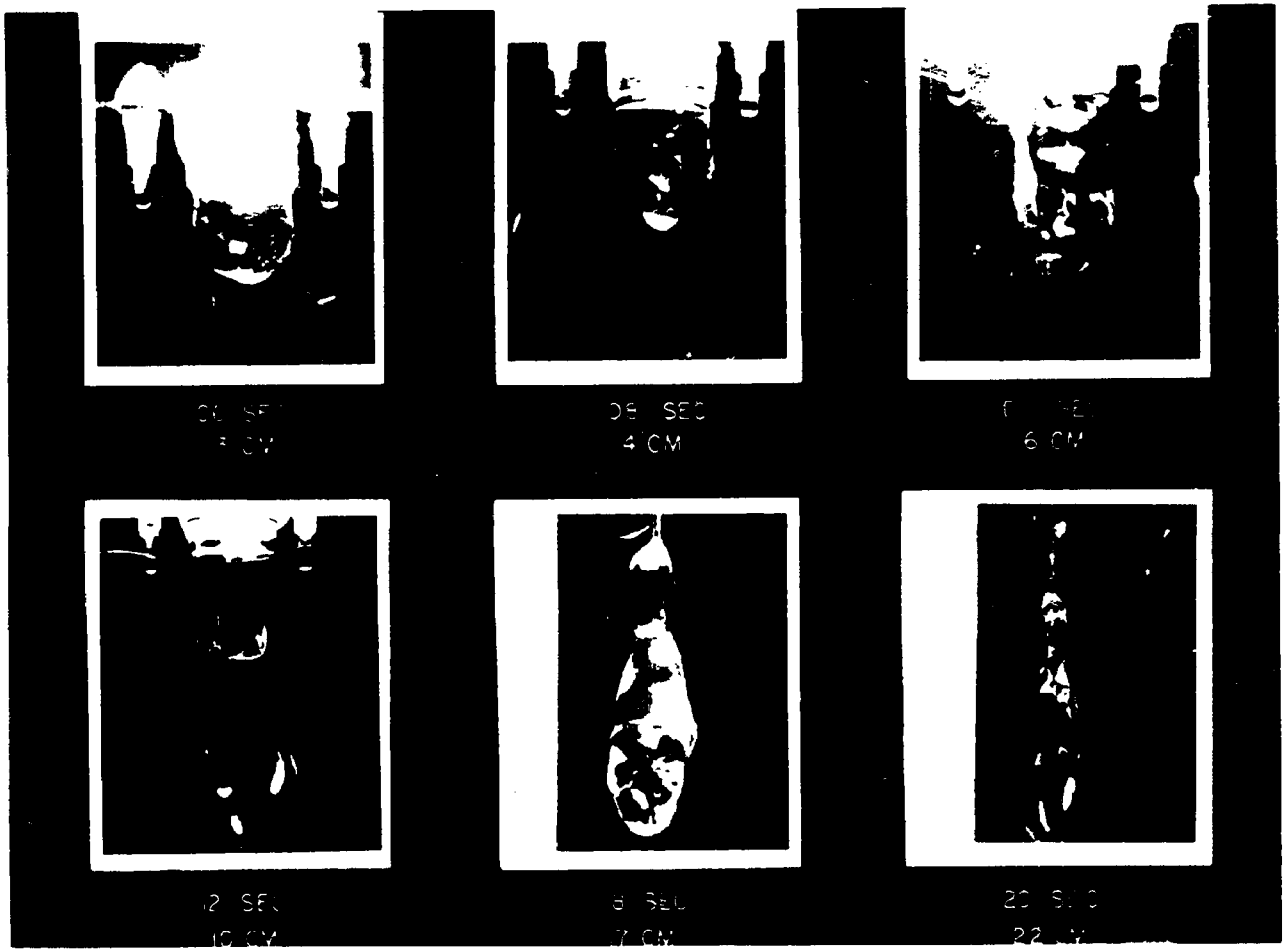


FIGURE 7

FLOW OF 25 ML OF LIQUID FROM CONTAINER — TIMES AND DISTANCES OF LIQUID FALL AFTER DIAPHRAGM RUPTURE

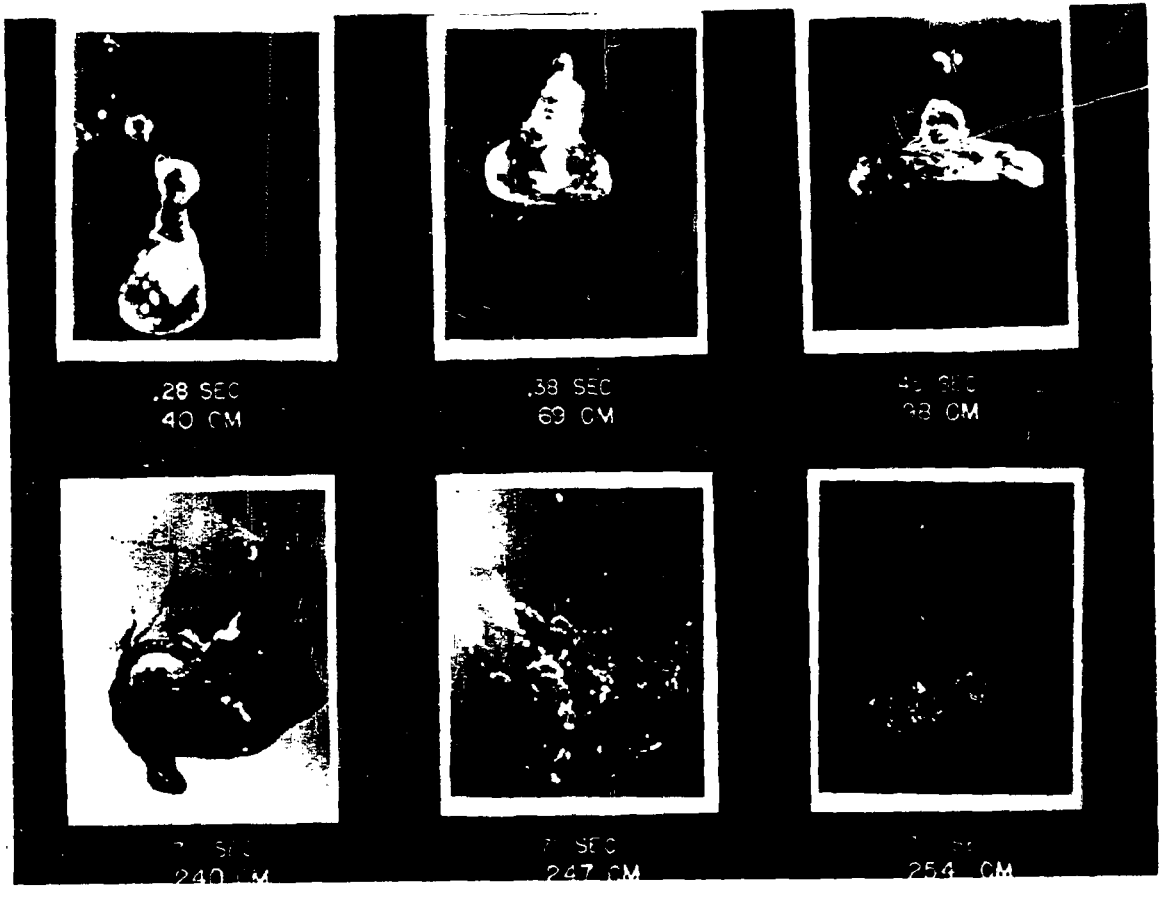


FIGURE 8

FREE-FALL BREAKUP OF 25 ML OF BULK LIQUID — INITIAL BREAKUP

(Times and distances of liquid fall after diaphragm rupture)

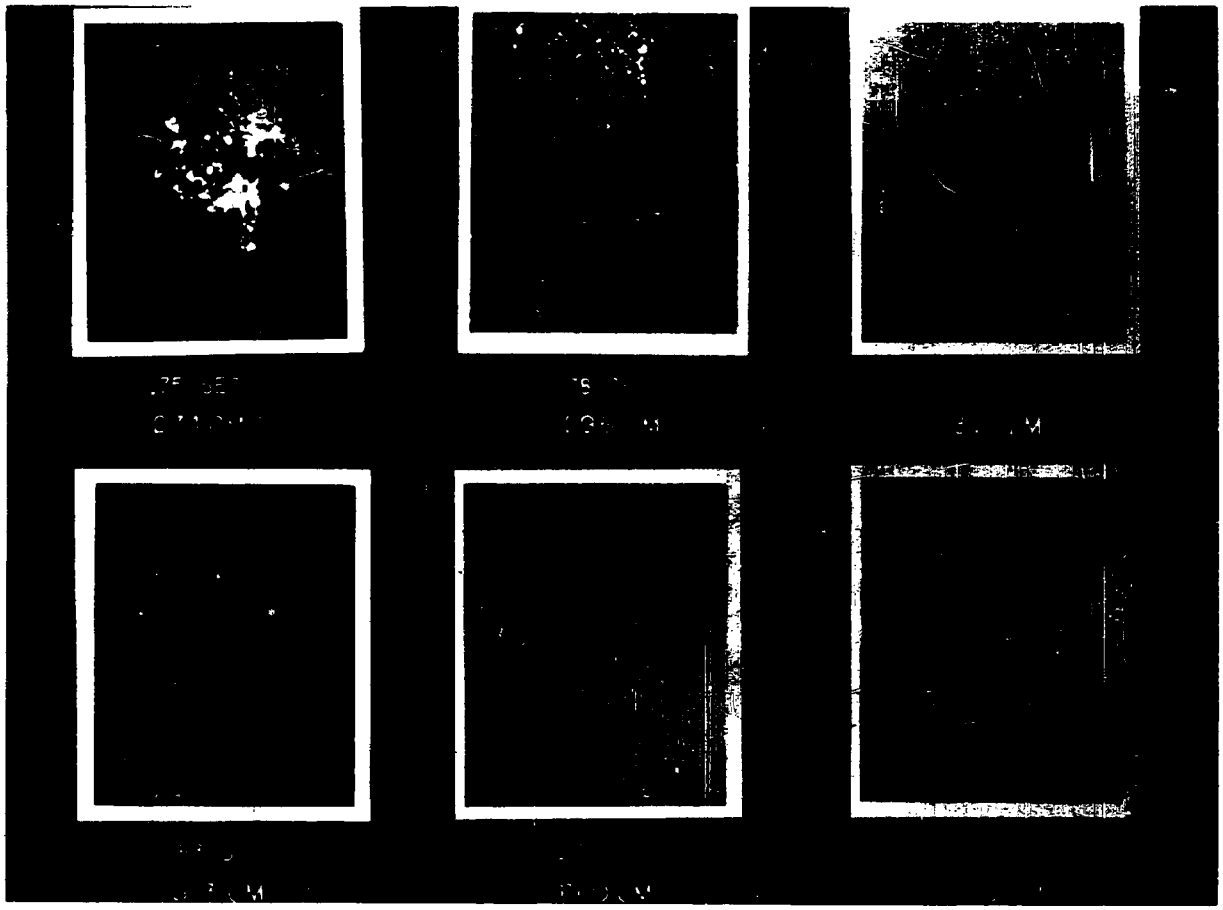


FIGURE 9

FREE-FALL BREAKUP OF 25 ML OF BULK LIQUID — SUBSEQUENT BREAKUP

(Times and distances of liquid after diaphragm rupture)

Figure 2 is a plot of the distance of fall after time of release as a function of time. The plot shows the first formation of droplets after a fall of 230 cm. The droplets fall at a lower velocity than the original liquid mass. The plot also shows that all of the original liquid mass has undergone some breakup after 5 m of fall and only droplets remain.

Figure 3 shows the relationship of velocity to distance after release of the liquid. The velocity at the point where the first bag broke was approximately 7 m/sec. The velocity at the point where all liquid was in droplet form was approximately 10 m/sec. At 8 m, the maximum height of these tests, a velocity of approximately 14 m/sec was reached.

B. Motion-Picture-Photography Method.

Figures 10, 11, and 12 are photographs of the free-fall breakup of 25-ml volumes of water, dibutyl phthalate, dibutyl phthalate containing 0.5% polyvinyl acetate polymer, and dibutyl phthalate containing 2.0% polyvinyl acetate polymer. The water contained 0.0025% fluorescein sodium dye to aid in photographing the liquid volume. The dye reduced the surface tension of the liquid from 69.0 to 60.3 dynes/cm. The effect of this surface-tension difference is discussed later.

The photographic resolution by the high-speed movie technique was not as good as the resolution by the still-photography method. This resolution difficulty prevented the determination of the time for complete flow of the liquid from the container. It is possible, however, to observe in figure 10 the times taken by the various liquids to fall 25 cm. The dibutyl phthalate containing 2.0% polymer had the highest fall velocity from the container. The dibutyl phthalate had a fall velocity of slightly less than this. The water and the dibutyl phthalate containing 0.5% polymer had the lowest fall velocities of the liquids tested. These photographs of 25-cm fall show the natural flow characteristics of the various liquids from the container.

The general over-all change of shape of the 25-ml volume of the liquid masses, the breakup of the masses, and the fall of the resulting breakup products is shown in figures 10, 11, and 12 as a function of distance and time. The two tests containing polyvinyl acetate demonstrated breakup by the bursting-bag or canopy-formation method. After breakup, these polymer solutions did not possess the fine particle sizes produced by the breakup of Newtonian liquids. The breakup products of the polymer solutions appeared to be large unstable particles undergoing further breakup. The resolution by the movie technique was barely sufficient for these observations. The breakup times and distances of the 25-ml volumes of the various liquids are shown in table 2.

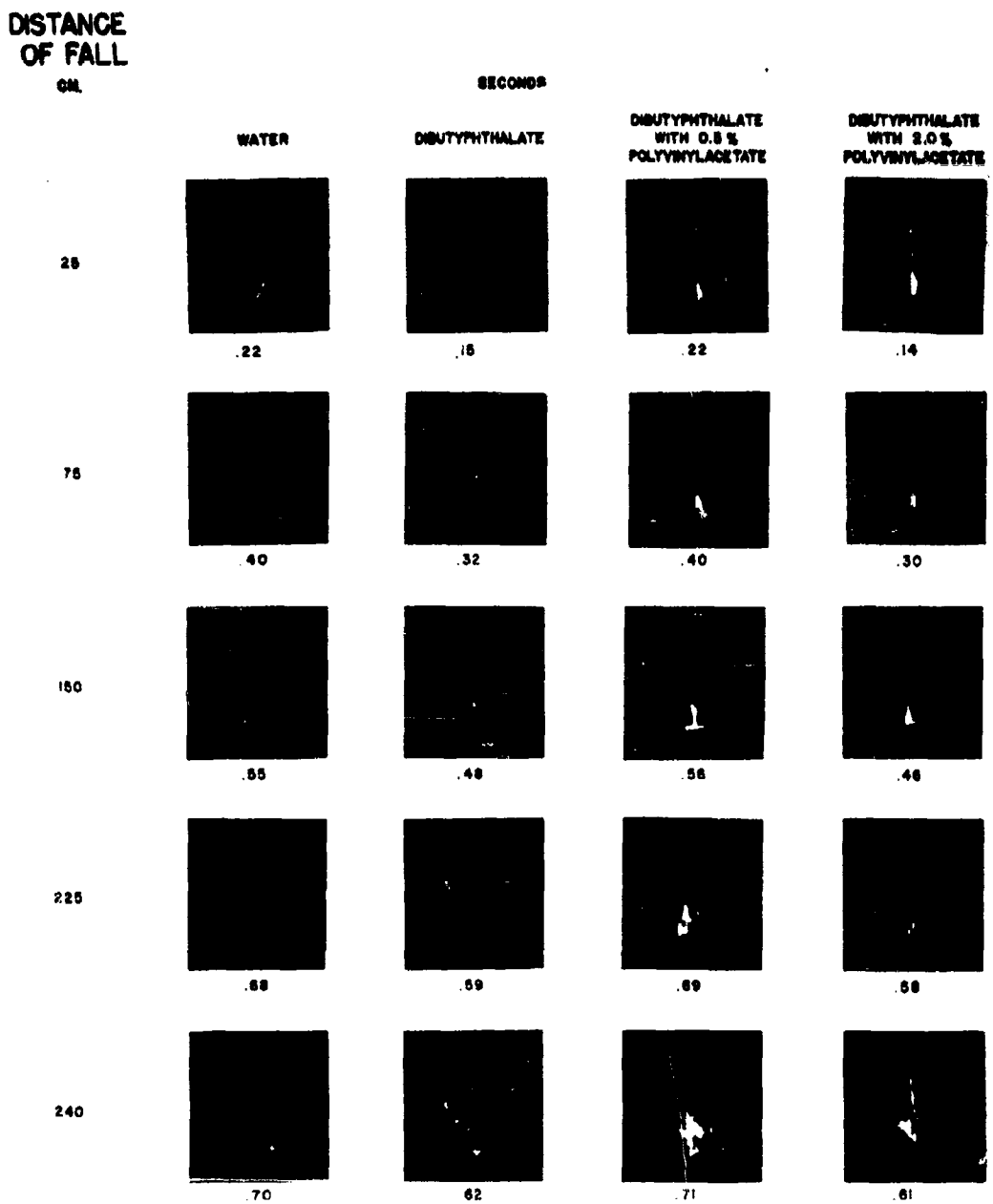


FIGURE 10

FREE-FALL BREAKUP OF 25 ML OF LIQUID

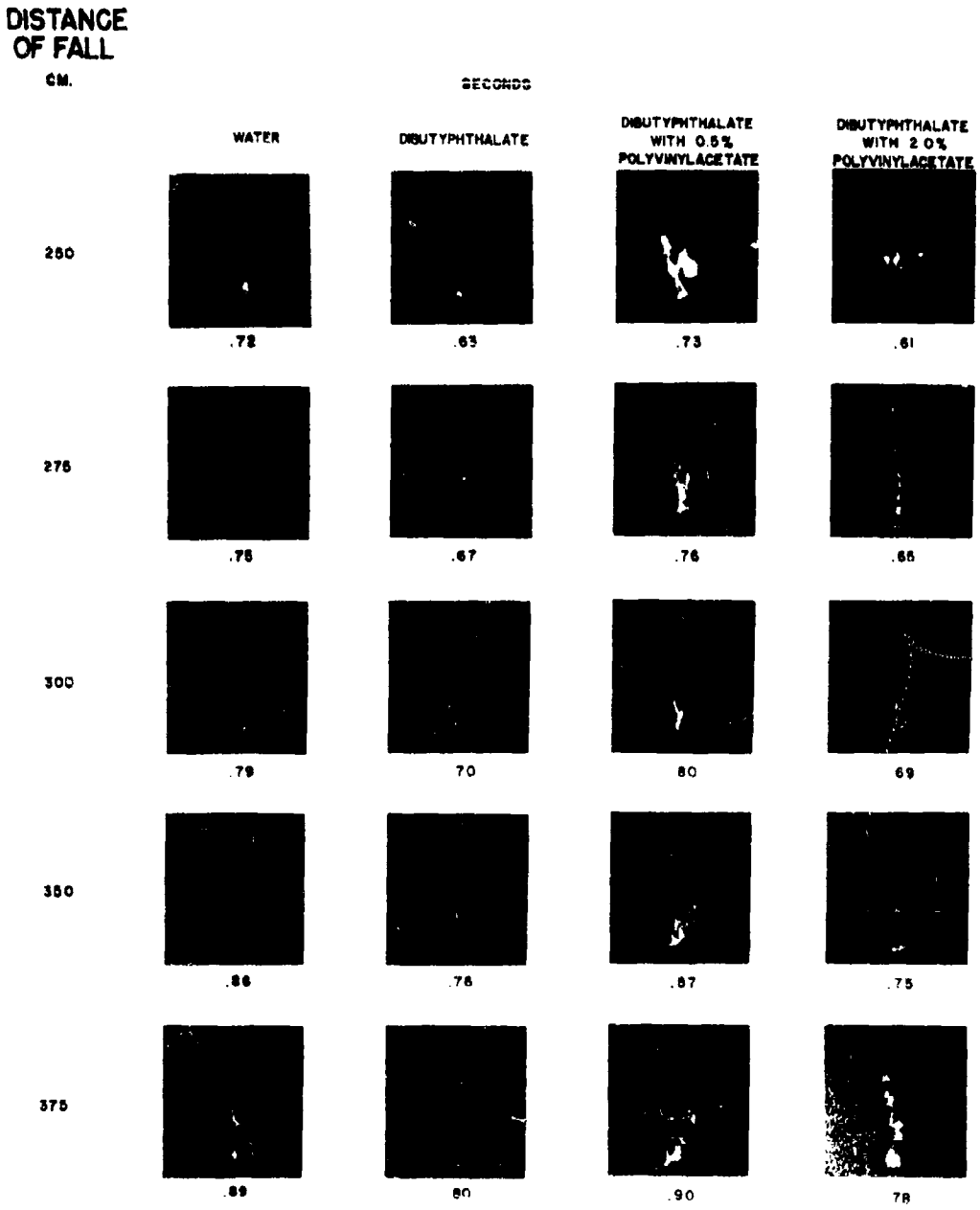


FIGURE 11

FREE-FALL BREAKUP OF 25 ML OF LIQUID

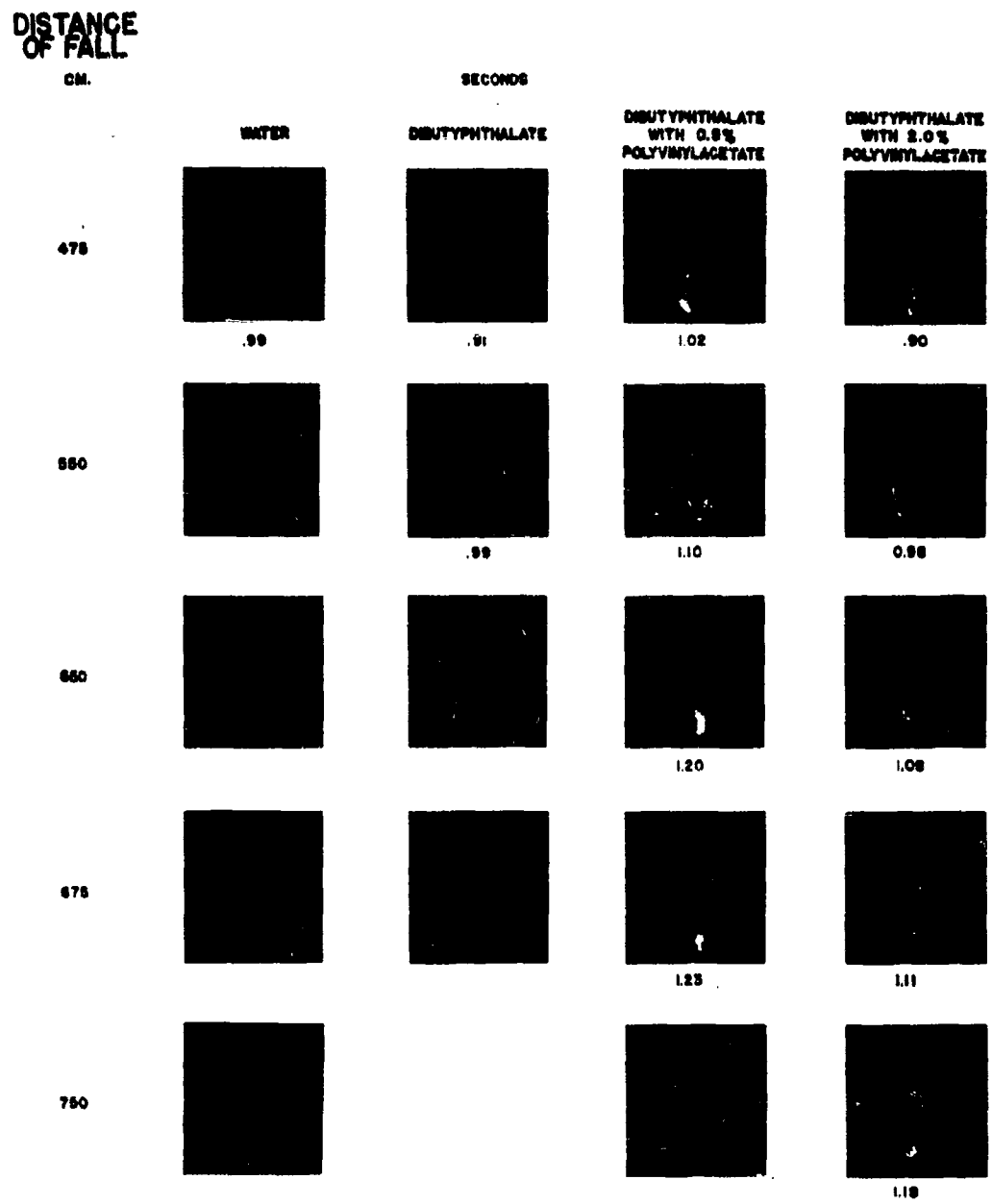


FIGURE 12

FREE-FALL BREAKUP OF 25 ML OF LIQUID

TABLE 2

BREAKUP TIMES AND DISTANCES OF LIQUIDS TESTED

Liquid composition	Breakup time	Breakup distance*
	sec	cm
Water containing traces of fluorescein sodium	0.77	290
Dibutyl phthalate	0.59	220
Dibutyl phthalate containing 0.5% polyvinyl acetate	0.75	260
Dibutyl phthalate containing 2.0% polyvinyl acetate	0.61	240

* Breakup distance is the distance from the point of liquid release to the point where the canopy or bag formed by the liquid is broken.

Figure 13 is a composite of three individual photographs showing the instantaneous release of 2.5 ℓ of water from a highly elastic rubber container. The left-hand portion of the photograph shows the rubber container in the process of being ruptured. This picture depicts the rapidity of withdrawal of the rubber container from the water mass. The curved section of the container with the white grainy appearance represents the area over which the contracting rubber has traveled after its rupture by a hypodermic needle. The white appearance of this section is caused by light scattering from the disturbed water surface. The dark curved line in the center of the white section represents the initial rupture of the container and indicates that the surface disturbance in this area is not as great as in the area immediately adjacent to it. The center portion of the photograph shows the 2.5-ℓ volume of water after the rubber has withdrawn itself completely from the liquid. In this photograph the liquid volume has fallen approximately 1 cm. The rupture of the rubber container, and its subsequent withdrawal from around the liquid volume because of its elasticity, did not appear to cause an uneven release of the water. The surface ripples, which appear to give the entire surface an unstable appearance, stabilize rather rapidly as shown in the right-hand portion of the photograph. In this picture the volume of water has fallen approximately 60 cm. The small droplets of water, which are separated from the

FIGURE 13

INSTANTANEOUS RELEASE OF 2.5 ℓ OF WATER FROM THIN HIGHLY ELASTIC RUBBER CONTAINER

main volume, were caused by the water trapped in the tail of the container at the time of rupture. In this portion of the photograph, a reduced image size was desirable to show the complete liquid mass in free fall and is a result of increasing the subject-to-camera distance.

The breakup of a 2.5-ℓ volume of water during 7 m of free fall is shown photographically in figure 14. The shape of the liquid during the initial and early stages of fall resembles those of the 25-ml liquid masses. The main process of breakup with this large quantity of liquid appears to be a stripping of the liquid from the outer surfaces of the mass. Some breakup is also taking place by the formation of thin sheets of liquid that break into ligaments. This type of breakup is seen best in the picture for 315-cm fall.

Figure 5 plots the relationship of the time and distance of fall of the liquid masses investigated. These liquid masses were 25-ml volumes of water containing a trace of fluorescein sodium dye, dibutyl phthalate, dibutyl phthalate containing 0.5% and 2.0% polyvinyl acetate, and a 2.5-ℓ volume of water containing a trace of fluorescein dye.

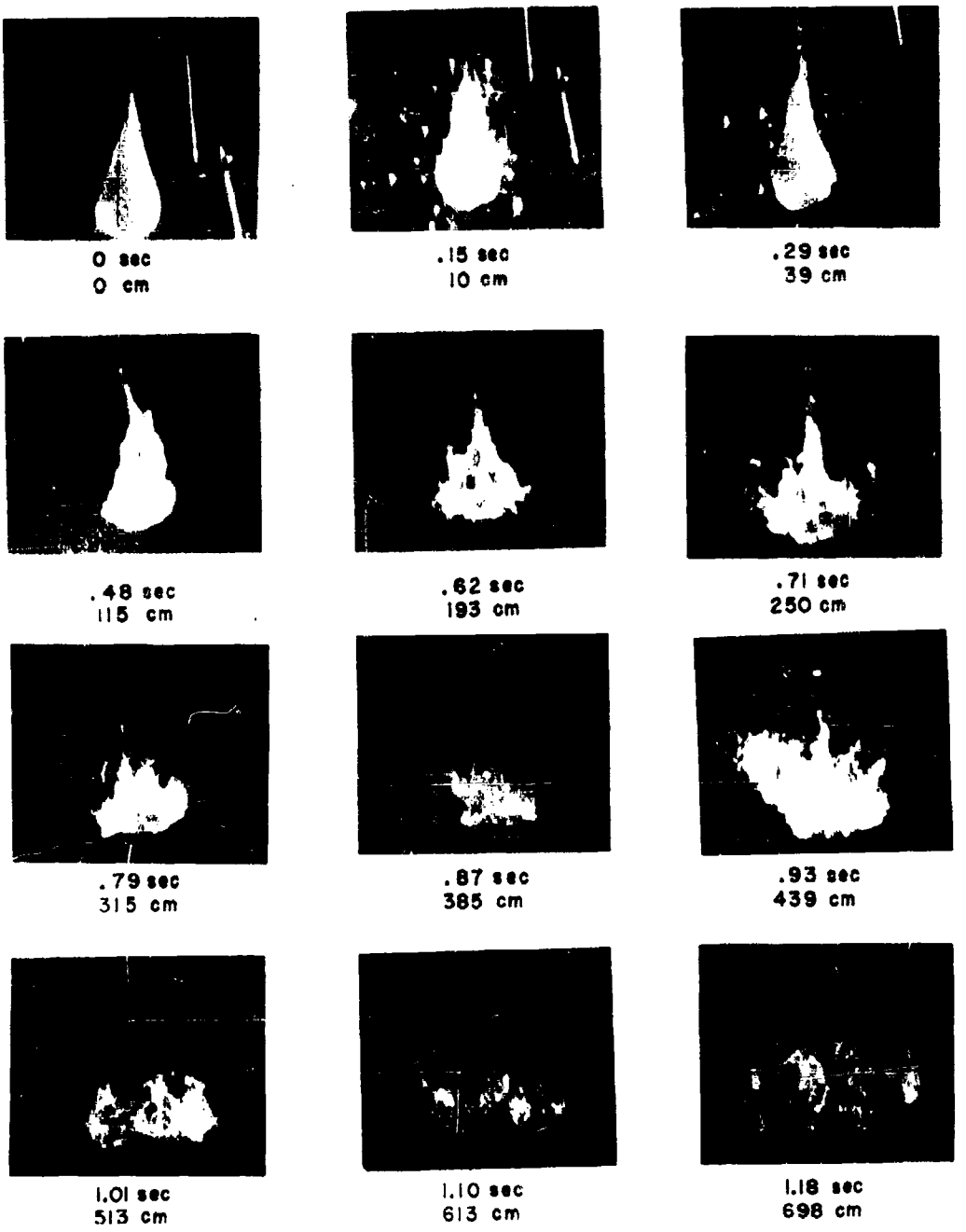
V. DISCUSSION.

It has been generally established in the open literature [6, 8, 9, 16] that waterdrops having a diameter larger than 7 mm are unstable and tend to break up when subjected to conditions of free fall. The terminal velocity of these 7-mm drops has not been well established; it has been generally accepted to be approximately 10 m/sec.

In this investigation with bulk liquid masses of water, which were considerably larger than the largest stable drop size mentioned above, the instability and subsequent breakup of these large masses were observed.

A. Still-Photography Method.

From tests conducted with 12.5-ml and 25-ml volumes of water, the original shape of the liquid might have been a factor in the liquid breakup. As the ratio of the length to the width of the liquid was increased, a portion of this liquid might have continued to fall after the main initial breakup pattern was completed and might in turn have formed a secondary breakup pattern. The breakup patterns formed by these unstable liquid masses are quite similar to the bursting-bag-type breakup described in the droplet experiments of Lane.[17] The velocity of the main liquid mass at the point of breakup was approximately 10 m/sec, approximately the same as the terminal velocity given for the unstable 7-mm drop mentioned previously. The velocity of the



TIMES AND DISTANCES OF FALL AFTER LIQUID RELEASE

FIGURE 14

FREE-FALL BREAKUP OF 2.5 ℓ OF WATER
(Times and distances of fall after liquid release)

mass of droplets increased to a value of approximately 14 m/sec after a fall of 8 m. This velocity increase would tend to indicate that not all of the liquid breakup was complete after 8 m of fall. It can be estimated from this velocity of 14 m/sec that the drops in the droplet mass are predominately larger than 7 mm in diameter. This can be visualized quite easily, remembering that the terminal velocity of drops smaller than 7 mm in diameter would be less than 10 m/sec. Since these droplets are traveling at a velocity of 14 m/sec, they must be unstable masses larger than 7 mm in diameter.

Since the breakup of 25 ml of water occurred by the bursting-bag process, and since smaller droplets also break up by this same process, as shown in the experiments of Lane,[17] the complete breakup of 25 ml of water is by the bursting-bag process. Furthermore, no droplets larger than 7 mm in diameter would result from the breakup of the 25 ml of water used if the test height had been great enough to allow complete breakup.

B. Motion-Picture-Photography Method.

In comparing the high-speed photographic methods of recording time and distance data, the time data obtained by the high-speed movie method were considerably more accurate than the time data obtained by the high-speed still-photography method. In comparing the quality of photographic resolution of the two methods, the degree of photographic resolution obtained by the still-photography method was far superior to that obtained by the high-speed movie method. It was estimated, however, that these factors introduced a combined error in distance and time measurements that did not exceed more than 10% in either experimental system.

Comparing the 25-ml water-breakup data obtained by both photographic methods, there appears to be very little difference in the breakup distances by either technique. The 25-ml water-breakup distance and time by still photography was 288 cm and 0.79 sec, and its breakup distance and time by the movie technique was 290 cm and 0.77 sec. This difference in time, as previously indicated, is most likely caused by the inaccuracy of the timing mechanism in the still-photographic method.

A slight difference in the data of the 25-ml water breakup obtained by the two methods would be expected as a result of the slight difference in surface tension. In the high-speed movie-photography method, the trace of fluorescein sodium added to the water lowered the surface tension of the water by about 10 dynes/cm. It is interesting to note that this

change in surface tension resulted in a change of the Weber number from 29 to 34. The Weber-number change is insignificant, since both values are well above the critical breakup Weber number of 10. Magarvey and Taylor[18] used a small quantity of aniline blue to assist in the photography of their water-breakup experiments. A trace of aniline blue lowered the surface tension of water slightly. Again a small surface-tension change appeared to produce an insignificant effect, since their breakup results were similar to those of Lane[17] and those shown in this study, even though Magarvey and Taylor were working with 15-mm drops. By comparison, liquids having a lower surface tension have an easily observable difference in breakup time and distance, as in the case of dibutyl phthalate, whose surface tension is 30.6 dynes/cm. A liquid having a lower surface tension than that of water will require less force to break it up and, consequently, will break up in a shorter time and distance. This would be expected and is demonstrated in the theoretical study of liquid breakup by the canopy-formation method.[3] In future studies, it should prove interesting to relate the Weber number of the drop at the time of breakup to the particle size of the final breakup products, since it appears that the higher the Weber number the more violent the breakup action. The various liquids tested resulted in different breakup times and distances as shown previously. The dibutyl phthalate containing 2.0% polyvinyl acetate fell the fastest because the polymer inhibited the deformation or flattening of the mass. This slow change in shape resulted in less air resistance and a faster fall velocity than any of the other liquids tested. Consequently, this increased velocity resulted in breakup that took place in a shorter time and distance than any of the other liquids tested. Even though this breakup was rapid, it appeared to be less complete than the breakup from any of the other liquids. It would be expected that this liquid would require the greatest height for complete breakup, and that the largest stable drop size would be greater than that of a waterdrop; i.e., in excess of 7 mm in diameter.

In the case of the dibutyl phthalate containing 0.5% polymer, examination of the results shows that the addition of the polymer caused the solution to have a slightly greater resistance to flow from the container than did the dibutyl phthalate itself. As in the case of the 2.0% polymer solution, this resistance to flow from the container appeared to be overcome by its slow change in shape or inhibition to deformation. Again in the case of the 0.5% polymer solution, the shape of the falling mass flattened much more quickly than did the 2.0% polymer solution. This slower exit velocity from the container and the rapid flattening of the 0.5% polymer solution mass that gave a greater air resistance resulted in a retarded rate of fall. The breakup time

and distance were greater than with the 2.0% polymer solution, since it took longer to attain the necessary force to cause the breakup. Again, this polymer solution did not appear to undergo complete breakup. It would also be expected that, because of its resistance to flow and shape change, a larger stable drop would result from this solution than with the dibutyl phthalate itself.

The water did not fall as fast as the dibutyl phthalate because of the difference in surface tension. The greater surface tension of the water keeps the liquid mass from attaining the shape necessary for breakup as quickly as the dibutyl phthalate. The resistance of the water mass to shape change results in a greater breakup distance and time than any of the other liquids tested.

It is interesting to note the change of shape of the 2.5-*l* volume and observe that the fall velocity over 7 m of fall of this large volume is roughly the same as the fall velocity of the 25-ml volume for the same distance. The orderly manner in which the breakup of the 2.5-*l* volume takes place shows that the whole breakup of the larger liquid volume could be estimated by a consideration of the mechanisms occurring during this breakup.

The height of the experimental setup could not be extended to more than 8 m because of the physical limitations involved. It was not possible to determine the height required for the complete breakup of 25 ml of water. This investigation indicated, however, that the larger the mass of liquid used, the greater the height necessary for complete breakup.

In general, the results of this study have indicated that the breakup of large liquid masses under free-fall conditions can be modified by altering certain physical factors. The factors that appear to have a modifying effect on the breakup of liquid masses under free fall include:

1. Surface tension; i.e., the lower the surface tension of the liquid the less the breakup distance.

2. The size of the liquid mass; i.e., the larger the mass the greater the breakup distance.

3. The initial shape of the liquid mass; i.e., when the liquid mass height is greater than its width, a greater distance of fall is needed to reach breakup conditions.

4. The velocity of the mass at the time of its release into the open atmosphere; i.e., the greater this imparted velocity the sooner disintegration will occur, and consequently, the breakup distance will be less.

5. The addition of a high-molecular-weight polymer to the liquid; i.e., liquids containing polymers did not undergo complete breakup.

The results of this study have also indicated that the predominant mode of breakup of a bulk-liquid mass subjected to free-fall conditions occurs by the bursting-bag or canopy-formation process. When the masses of liquid are very large, other mechanisms of breakup take place; namely, liquid-film formation and surface stripping. During the final stages of breakup, after smaller masses have broken away from the main mass, breakup again takes place by the canopy-formation method.

As a result of this study, it is felt that the dissemination of a large bulk of liquid by release from some altitude above the ground and its free-fall or accelerated-fall conditions merit further study where large drops of liquid are desired. Large drops are desirable in certain instances since they have a rapid settling rate. The experimental results and high-speed photographs indicate that larger drops should result when a polymer of high molecular weight is added to the liquid. This is also the case for liquid breakup under high acceleration conditions.[22] It is expected that the size of the breakup products from a liquid containing a high-molecular-weight polymer would be a function of the force acting to break up the liquid; that is, the velocity difference between the liquid and the air, and the molecular weight of the polymer used and its concentration.

VI. CONCLUSIONS

The following conclusions were reached:

1. The predominant mode of breakup of a bulk-liquid mass subjected to free-fall conditions occurs by the bursting-bag or canopy-formation process.

2. Certain physical factors in the liquid mass, such as surface tension, size, initial shape, velocity at the time of release, and concentration of high-molecular-weight polymer, have a modifying effect on its breakup.

LITERATURE CITED

1. Armour Research Foundation. Contract DA-18-108-CML-5507, Report 13. ETF 100.4-7/13. Analysis of Selected Dissemination and Design Problems. December 1955. SECRET Report.

2. Rosinski, J., et al. Armour Research Foundation. Contract DA-18-108-CML-5507, ETF 100.4-7/Task XII. Production of Toxic Rain. August 1957. SECRET Report.

3. Garcia, J. D., and Wilcox, J. D. CRDLSP 1-30 (in press). Breakup of Liquid Masses in Free Fall by Canopy Formation. UNCLASSIFIED Report.

4. Lenard, P. Uber Regen. Meteorol. Z. 21, 248 (1904).

5. Schmidt, W. Eine unmittelbare Bestimmung der Fallgeschwindigkeit von Regentropfen. Ber. Akad. Wiss. Wien 118, 71 (1909).

6. Laws, J. O. Measurement of the Fall-Velocity of Water-Drops and Rain-Drops. Trans. Am. Geophys. Union 22, 709 (1941).

7. Gunn, R., and Kinzer, G. D. The Terminal Velocity of Fall for Water Droplets in Air. J. Meteorol. 6, 243 (1949).

8. Davies, C. N. (quoted by O. G. Sutton). No. 40, Meteorological Research Paper. Meteorology Office, London. 1942.

9. Best, A. C. The Size Distribution of Raindrops. Quart. J. Roy. Meteorol. Soc. 76, 16 (1950).

10. Repp, C. W., and Magrum, S. J. CWL Technical Memorandum 5-5. Empirical Equations for Terminal Settling Velocities. August 1958.

11. Mason, B. J. The Physics of Clouds. p 436. Clarendon Press, London. 1957.

12. Drinker, P., and Hatch, T. Industrial Dust. First ed. p 6. McGraw-Hill Book Company, Inc., New York. 1936.

13. Magono, C. On the Shape of Water Drops Falling in Stagnant Air. J. Meteorol. 11, 77 (1954).

14. McDonald, J. E. The Shape and Aerodynamics of Large Raindrops. J. Meteorol. 11, 478 (1954).

15. Gunn, R. Mechanical Resonance in Freely Falling Raindrops. J. Geophys. Research 54, 383 (1949).

16. Blanchard, D. C. The Behavior of Water Drops at Terminal Velocity in Air. Trans. Am. Geophys. Union 31, 836 (1950).

17. Lane, W. R. Shatter of Drops in Streams of Air. Ind. Eng. Chem. 43, 1312 (1951).

18. Magarvey, R. H., and Taylor, B. W. Free Fall Breakup of Large Drops. J. Appl. Phys. 27, 1129 (1956).

19. Hughes, R. R., and Gilliland, E. R. The Mechanics of Drops. Chem. Eng. Progr. 48, 497 (1952).

20. Hinze, J. O. Forced Deformations of Viscous Liquid Globules. Appl. Sci. Research A1, 263 (1948).

21. Merrington, A. C., and Richardson, E. G. The Break-Up of Liquid Jets. Proc. Phys. Soc. (London) 59, Part 1, 1-13 (1947).

22. Wilcox, J. D., et al. CWLR 2309. The Effect of Polymeric Modifiers on the Breakup of Drops by High-Velocity Air Streams. September 1959. UNCLASSIFIED Report.

UNCLASSIFIED

AD_____ Accession No._____
Physicochemical Research Division, U. S. Army Chemical Research and Development Laboratories, Army Chemical Center, Maryland

FREE-FALL BREAKUP OF BULK LIQUIDS - Alan B. Palmer, Joseph V. Pistritto, and James D. Wilcox

CRDLR 3085, September 1961
Task 4C08-03-016-10, UNCLASSIFIED REPORT

The mechanism and characteristics of the breakup of large bulk masses of liquid under free-fall conditions were investigated. The breakup of 12.5-ml to 2.5-*l* masses of water and 25-ml masses of dibutyl phthalate and dibutyl phthalate solutions containing 0.5% and 2.0% polyvinyl acetate by the bursting-bag or canopy-formation process is described. Their breakup was recorded by high-speed photographic techniques. The influence of certain physical parameters such as surface tension, size and geometrical shape of the mass, mass velocity at time of release, and the addition of small quantities of high-molecular-weight polymers in controlling the bulk-liquid breakup is shown.

1. Liquids, dissemination and breakup of
2. Drops, breakup of
3. Aerosols
4. Velocity, free-fall breakup

UNCLASSIFIED

UNCLASSIFIED

AD_____ Accession No._____
Physicochemical Research Division, U. S. Army Chemical Research and Development Laboratories, Army Chemical Center, Maryland

FREE-FALL BREAKUP OF BULK LIQUIDS - Alan B. Palmer, Joseph V. Pistritto, and James D. Wilcox

CRDLR 3085, September 1961
Task 4C08-03-016-10, UNCLASSIFIED REPORT

The mechanism and characteristics of the breakup of large bulk masses of liquid under free-fall conditions were investigated. The breakup of 12.5-ml to 2.5-*l* masses of water and 25-ml masses of dibutyl phthalate and dibutyl phthalate solutions containing 0.5% and 2.0% polyvinyl acetate by the bursting-bag or canopy-formation process is described. Their breakup was recorded by high-speed photographic techniques. The influence of certain physical parameters such as surface tension, size and geometrical shape of the mass, mass velocity at time of release, and the addition of small quantities of high-molecular-weight polymers in controlling the bulk-liquid breakup is shown.

1. Liquids, dissemination and breakup of
2. Drops, breakup of
3. Aerosols
4. Velocity, free-fall breakup

UNCLASSIFIED

UNCLASSIFIED

AD_____ Accession No._____
Physicochemical Research Division, U. S. Army Chemical Research and Development Laboratories, Army Chemical Center, Maryland

FREE-FALL BREAKUP OF BULK LIQUIDS - Alan B. Palmer, Joseph V. Pistritto, and James D. Wilcox

CRDLR 3085, September 1961
Task 4C08-03-016-10, UNCLASSIFIED REPORT

The mechanism and characteristics of the breakup of large bulk masses of liquid under free-fall conditions were investigated. The breakup of 12.5-ml to 2.5-*l* masses of water and 25-ml masses of dibutyl phthalate and dibutyl phthalate solutions containing 0.5% and 2.0% polyvinyl acetate by the bursting-bag or canopy-formation process is described. Their breakup was recorded by high-speed photographic techniques. The influence of certain physical parameters such as surface tension, size and geometrical shape of the mass, mass velocity at time of release, and the addition of small quantities of high-molecular-weight polymers in controlling the bulk-liquid breakup is shown.

1. Liquids, dissemination and breakup of
2. Drops, breakup of
3. Aerosols
4. Velocity, free-fall breakup

UNCLASSIFIED